

Scattering and radiative properties of semi-external versus external mixtures of different aerosol types

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Received 15 November 2003; accepted 15 December 2003

Abstract

The superposition *T*-matrix method is used to compute the scattering of unpolarized light by semi-external aerosol mixtures in the form of polydisperse, randomly oriented two-particle clusters with touching components. The results are compared with those for composition-equivalent external aerosol mixtures, in which the components are widely separated and scatter light in isolation from each other. It is concluded that aggregation is likely to have a relatively weak effect on scattering and radiative properties of two-component tropospheric aerosols and can be replaced by the much simpler external-mixture model in remote sensing studies and atmospheric radiation balance computations. Published by Elsevier Ltd.

Keywords: Aerosols; Scattering; Optical cross sections; Single-scattering albedo; Asymmetry parameter; Scattering matrix; Remote sensing; Atmospheric radiation

1. Introduction

Different kinds of tropospheric aerosols (e.g., dust, carbonaceous, and sulfate particles) can be suspended in air both in the form of external mixtures (different aerosol particles are separated by distances much greater than their sizes and scatter light independently of each other), semi-external mixtures (two or more aerosol particles are in physical contact and form an aggregate), and internal mixtures (one or several small aerosol particles are imbedded in a larger host particle) (see Fig. 1). For example, Podzimek [1] found that in a polluted urban-marine environment haze elements can exist as insoluble, carbonaceous

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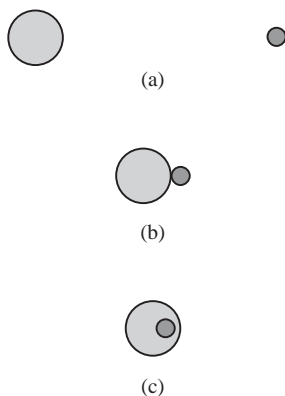


Fig. 1. (a) External, (b) semi-external, and (c) internal particle mixtures.

particles on or in larger nonabsorbing droplets. Significant aggregation of soot and/or sulfate with mineral dust is common when dust from the Chinese interior mixes with pollution plumes from major industrial cities. Since aerosol particles forming an aggregate are in the near field zone of each other, the scattering and radiative properties of semi-external aerosol mixtures can differ from those of composition-equivalent external mixtures. These potential differences may influence the results of remote sensing studies of tropospheric aerosols and calculations of the direct aerosol forcing of climate (see, e.g., [2] and references therein) and, therefore, must be accurately evaluated using theoretical calculations and/or laboratory measurements [3–5].

In this paper we calculate the optical cross sections and the elements of the Stokes scattering matrix of semi-external two-component mixtures of carbonaceous, dust, and sulfate aerosols—the three types of aerosols which are believed to have a significant effect on the earth’s climate—and compare them with those of equivalent external mixtures computed with the standard Lorenz–Mie theory [6,7]. This comparison is used to derive conclusions about the likely effect of aggregation on the scattering and radiative properties of multi-component aerosols.

2. Computations

Important single-scattering characteristics of randomly oriented particles with a plane of symmetry are the ensemble-averaged scattering, C_{sca} , and extinction, C_{ext} , cross sections and the elements of the normalized scattering matrix $\mathbf{F}(\Theta)$, where Θ is the scattering angle [7]. In the standard $\{I, Q, U, V\}$ representation of polarization, the scattering matrix has the well-known block-diagonal form [6,7],

$$\mathbf{F}(\Theta) = \begin{bmatrix} a_1(\Theta) & b_1(\Theta) & 0 & 0 \\ b_1(\Theta) & a_2(\Theta) & 0 & 0 \\ 0 & 0 & a_3(\Theta) & b_2(\Theta) \\ 0 & 0 & -b_2(\Theta) & a_4(\Theta) \end{bmatrix}, \quad (1)$$

so that only eight elements of $\mathbf{F}(\Theta)$ are nonzero and only six of them are independent. The (1, 1) element of the scattering matrix $a_1(\Theta)$ is traditionally called the phase function and satisfies the normalization

condition

$$\frac{1}{2} \int_0^\pi d\Theta \sin \Theta a_1(\Theta) = 1. \quad (2)$$

Additional useful quantities are the ensemble-averaged absorption cross section, $C_{\text{abs}} = C_{\text{ext}} - C_{\text{sca}}$, the single-scattering albedo, $\varpi = C_{\text{sca}}/C_{\text{ext}}$, and the asymmetry parameter defined as

$$g = \frac{1}{2} \int_0^\pi d\Theta \sin \Theta a_1(\Theta) \cos \Theta. \quad (3)$$

The computations reported here were performed at a visible wavelength of $\lambda = 0.628 \mu\text{m}$ assuming that each aerosol mode is represented by the power law size distribution of the following type [7,8]:

$$n(r) = \begin{cases} \frac{2r_1^2 r_2^2}{r_2^2 - r_1^2} r^{-3}, & r_1 \leq r \leq r_2, \\ 0 & \text{otherwise.} \end{cases} \quad (4)$$

The effective radius r_{eff} and effective variance v_{eff} of the size distribution are defined as

$$r_{\text{eff}} = \frac{1}{\langle G \rangle} \int_{r_1}^{r_2} dr n(r) r \pi r^2, \quad (5)$$

$$v_{\text{eff}} = \frac{1}{\langle G \rangle r_{\text{eff}}^2} \int_{r_1}^{r_2} dr n(r) (r - r_{\text{eff}})^2 \pi r^2, \quad (6)$$

where

$$\langle G \rangle = \int_{r_1}^{r_2} dr n(r) \pi r^2 \quad (7)$$

is the average area of the particle geometrical projection [7,8]. Dust aerosols are usually characterized by a broad range of effective radii from about $0.2 \mu\text{m}$ to greater than $5 \mu\text{m}$. The most common dust mode occurring at a large distance from the source is around $1 \mu\text{m}$ in radius. The effective radius of sulfate particles ranges from less than 0.1 to larger than $0.5 \mu\text{m}$ and depends on relative humidity. Soot aerosols are usually fine particles with effective radii around $0.1 \mu\text{m}$.

In our computations, we have used two types of aerosol particle mixture. Mixture 1 is composed of dust-like particles with an effective radius of $1 \mu\text{m}$ and an equal number of sulfate particles. The effective radius of the sulfate component was set at the following four representative values: 0.1 , 0.2 , 0.5 , and $1 \mu\text{m}$. Mixture 2 consists of equal numbers of sulfate particles with an effective radius of $0.5 \mu\text{m}$ and soot aerosols. The effective radius of soot particles was set at 0.05 , 0.1 , 0.2 , and $0.5 \mu\text{m}$. We used the following values of the relative refractive index for the three different aerosol species: $1.53 + 0.008i$ for dust, 1.44 for sulfates, and $1.75 + 0.435i$ for soot [9]. The effective variance for all aerosol types was fixed at 0.2 , thereby representing a moderately wide size distribution. Although solid aerosol particles should be presumed to have nonspherical shapes, our main interest here is in evaluating the potential effect of aggregation on light scattering. Therefore, for the sake of simplicity, we have assumed that all three aerosol species consist of spherical particles.

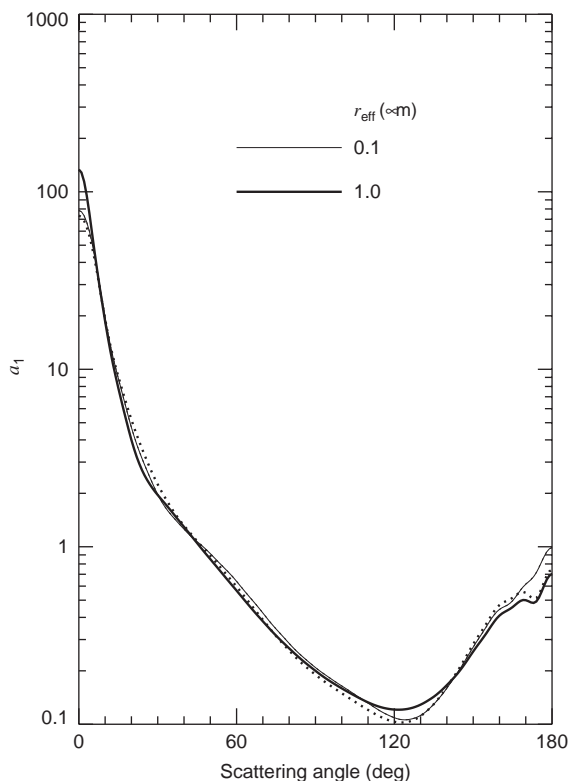


Fig. 2. Phase function versus scattering angle for dust–sulfate semi-external (solid curves) and external (dotted curves) mixtures. The effective radius of dust particles is fixed at 1 μm . The thin and thick curves, respectively, show the results for the smallest ($r_{\text{eff}} = 0.1 \mu\text{m}$) and the largest ($r_{\text{eff}} = 1 \mu\text{m}$) sulfate aerosol mode considered in this paper.

We calculated the scattering of unpolarized incident light by polydisperse, randomly oriented two-sphere clusters with touching components and compared the results with the results of Lorenz–Mie calculations assuming that the same component particles were mixed externally and acted as independent scatterers. In our computations for two-sphere clusters, we used a highly efficient code developed by Mishchenko and Mackowski [10] and publicly available at <http://www.giss.nasa.gov/~crmim>. This code is based on the T -matrix solution of Maxwell’s equations and precisely computes the scattering of light by randomly oriented two-sphere clusters with sizes comparable to and larger than the wavelength. The efficiency of the methods is the result of combining the power of the superposition approach in treating light scattering by composite particles [11,12] and the analyticity of the T -matrix formulation in application to randomly oriented nonspherical scatterers [13]. The main idea of the method is to employ the superposition approach to calculate the T matrix of a bisphere in the natural coordinate system with the z -axis connecting the centers of the component spheres and then to use this T matrix in an analytical procedure to directly compute the orientation-averaged optical cross sections and the elements of the scattering matrix for randomly oriented bispheres. The analytical averaging over orientations makes this approach much faster than that based on the standard numerical averaging and, thus, suitable for

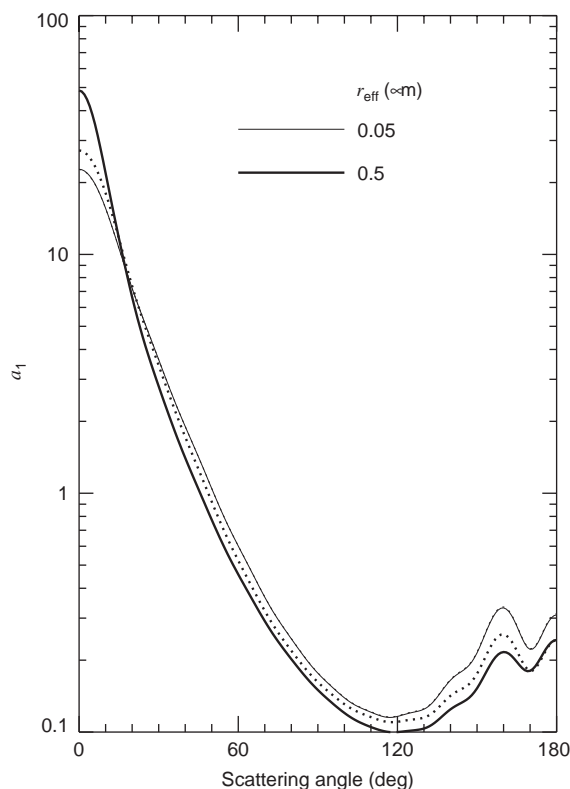


Fig. 3. As in Fig. 2, but for sulfate–soot mixtures. The effective radius of sulfate particles is fixed at $0.5\ \mu\text{m}$. The thin and thick curves, respectively, show the results for the smallest ($r_{\text{eff}} = 0.05\ \mu\text{m}$) and the largest ($0.5\ \mu\text{m}$) soot aerosol mode considered in this paper.

computations for realistic size distributions. The Lorenz–Mie code used is described in [7] and is also available at <http://www.giss.nasa.gov/~crmim>. The numerical data are summarized in Figs. 2, 3, 4, 5 and Tables 1 and 2 and are discussed in the following section.

3. Discussion

Figs. 2 and 3 illustrate the differences between the phase functions for semi-external (solid curves) and external (dotted curves) aerosol mixtures. One can see that the relative phase-function differences are rather small and, on average, do not exceed 20%. A notable exception is the exact forward-scattering direction, where the interference effects result in a significant enhancement of the aggregate phase functions [14]. The phase-function differences are especially small when the effective radius of one component is much smaller than that of the other, in which cases the thin solid and dotted curves are almost (or even completely) indistinguishable. This obviously happens because the contribution of the smaller component to the total scattered signal becomes negligibly small.

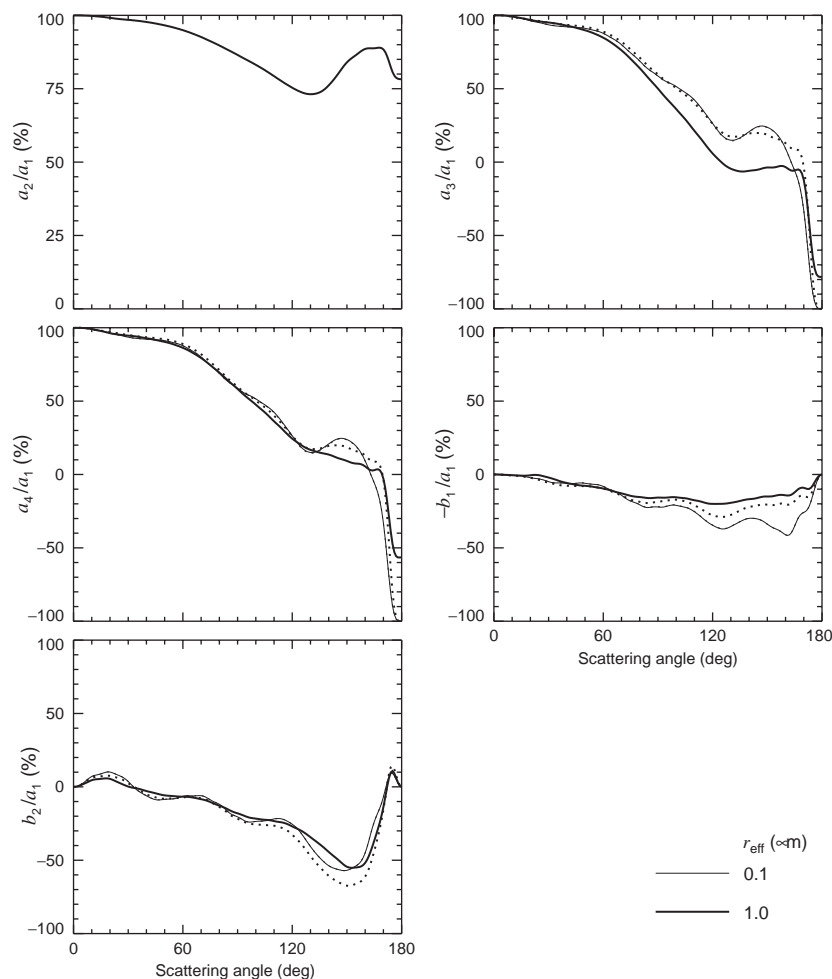


Fig. 4. As in Fig. 2, but for scattering-matrix element ratios.

The differences in the other scattering-matrix elements are also surprisingly small (Figs. 4 and 5). The only clear indications of the fact that two-sphere aggregates are nonspherical particles are the deviation of the ratio $a_2(\Theta)/a_1(\Theta)$ from unity and the deviation of the ratios $a_3(180^\circ)/a_1(180^\circ)$ and $a_4(180^\circ)/a_1(180^\circ)$ from -1 (cf. [7,14]).

Tables 1 and 2 demonstrate that the differences in the integral photometric characteristics (the ensemble averaged extinction, scattering, and absorption cross sections, the single-scattering albedo, and the asymmetry parameter) between the semi-external and the composition-equivalent external mixtures do not exceed 15% and are often much smaller, particularly for C_{abs} , ϖ , and g . Owing to mutual shadowing, the scattering cross sections of the semi-externally mixed aerosols are always smaller than those of their externally mixed counterparts, whereas the absorption cross section hardly changes.

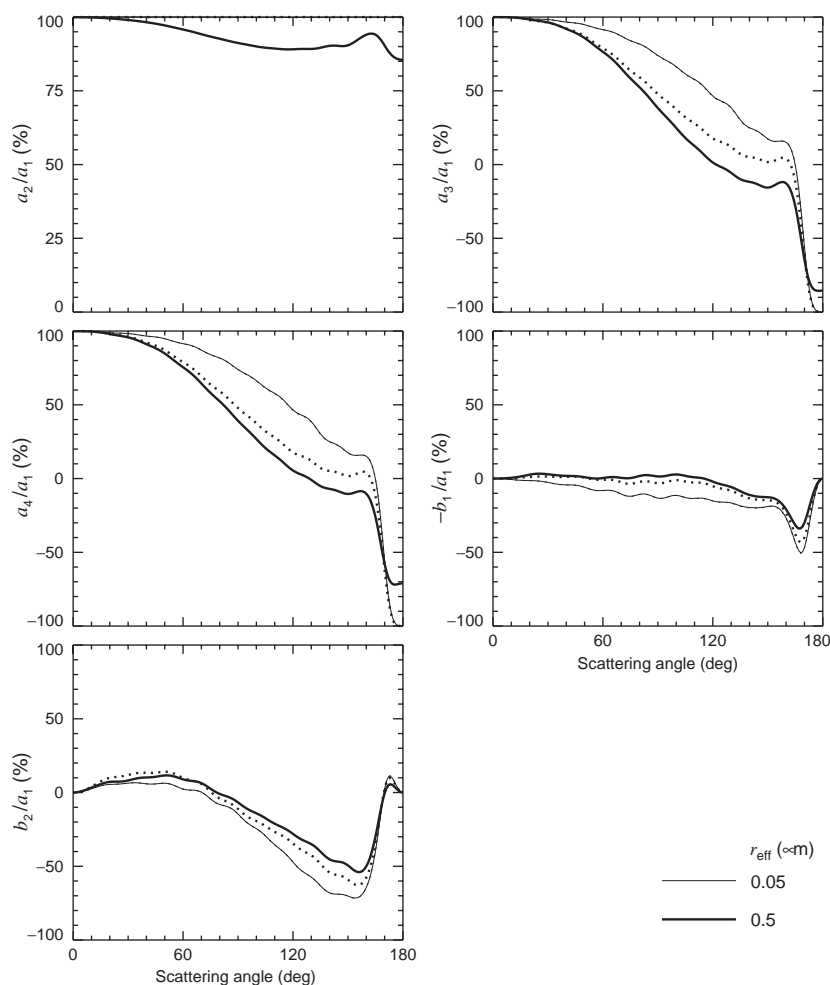


Fig. 5. As in Fig. 3, but for scattering-matrix element ratios.

The small differences in the single-scattering albedo and the asymmetry parameter in combination with the small phase function and polarization differences suggest that aggregation is likely to have a relatively weak effect on remote sensing retrievals of the aerosol physical characteristics and computations of the aerosol direct radiative forcing. This is especially true when one of the aerosol components is much larger than the other and, thereby, dominates the total optical characteristics of the mixture. It is also quite obvious that the semi-external mixing does not cause as pronounced an enhancement of the black carbon absorption as that caused by internal mixing [3]. As a consequence, it appears to be rather safe to use the much simpler external mixture model in radiative transfer computations irrespective of the actual form of mixing.

The analysis of this paper is limited to simple two-particle aggregates. Since more complex aerosol aggregates can also be encountered in the atmosphere, we plan to extend this study by performing

Table 1

Optical characteristics of dust–sulfate particle mixtures^a

r_{eff} (μm)	0.1		0.2		0.5		1	
	SE ^b	E	SE	E	SE	E	SE	E
C_{ext} (μm^2)	4.540	4.542	4.578	4.635	5.319	5.794	8.309	9.361
C_{sca} (μm^2)	3.952	3.955	3.988	4.048	4.727	5.207	7.716	8.774
C_{abs} (μm^2)	0.588	0.587	0.590	0.587	0.592	0.587	0.593	0.587
ϖ	0.871	0.871	0.871	0.873	0.889	0.899	0.929	0.937
g	0.712	0.712	0.711	0.712	0.716	0.715	0.726	0.720

^aThe effective radius of dust particles is fixed at 1 μm .^b“SE” denotes semi-external mixing and “E” denotes external mixing.

Table 2

Optical characteristics of sulfate–soot aerosol mixtures^a

r_{eff} (μm)	0.05		0.1		0.2		0.5	
	SE ^b	E	SE	E	SE	E	SE	E
C_{ext} (μm^2)	1.259	1.259	1.272	1.282	1.367	1.426	2.176	2.423
C_{sca} (μm^2)	1.256	1.257	1.255	1.266	1.269	1.332	1.563	1.822
C_{abs} (μm^2)	0.003	0.002	0.017	0.016	0.098	0.094	0.613	0.601
ϖ	0.998	0.998	0.986	0.988	0.928	0.934	0.718	0.752
g	0.722	0.721	0.726	0.720	0.737	0.717	0.780	0.751

^aThe effective radius of sulfate particles is 0.5 μm .^b“SE” denotes semi-external mixing and “E” denotes external mixing.

calculations using the code described in [15] and applicable to randomly oriented clusters with three or more components.

Acknowledgements

This research was supported by the NASA Radiation Sciences Program managed by Donald Anderson.

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